Phonon profiles in superconducting YNi_2B_2C and $LuNi_2B_2C$

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We performed a systematic inelastic neutron scattering, between 4 and 620 K, of the profiles of the anomalous acoustic and optic phonon observed in both compounds in the vicinity of $\vec{\xi}_m \cong (0.5\ 0\ 0)$. Above T_c we find that, in both compounds, the frequencies of the acoustic modes decrease with decreasing temperature and there is intensity transfer from the upper (optic) to the lower (acoustic) mode. The frequencies of the optic modes of the Y compound, on the other hand, decrease slower than those of the Lu compound as the temperature decreases towards T_c and, as a result, the two branches approach each other in the Lu compound, whereas they separate in the Y compound. In both compounds the observed phonon profiles above T_c can be described satisfactorily in terms of a simple couple-mode model. Comparison of the phonon spectra of the two compounds below T_c demonstrates that the sharp peak at approximately 4 meV with a broad shoulder at the higher-energy side, observed in both compounds, arises from the change in the acoustic phonon profile upon entering the superconducting state. [S0163-1829(99)07641-9]

I. INTRODUCTION

The compounds of the RNi_2B_2C family (R=rare earth) are ideal for a detailed study of the competition between magnetic ordering, superconductivity, and lattice instabilities.[1–3] Detailed neutron-scattering experiments[4–7] on the superconducting Lu and Y compounds demonstrated the dramatic softening of the $\Delta_4[\xi 00]$ acoustic modes in the vicinity of (0.500), which is practically the same as the nesting vector $\vec{\xi}_m$ obtained by band theoretical calculations.[3] The softening is so significant that it was also observed in the phonon density of states[8] and the point-contact spectra[9] of these compounds. This is an unusual type of softening involving the acoustic and lowest-lying optic $\Delta_4[\xi 00]$ branches, which cannot cross by symmetry (see Fig. 1). These two branches are strongly coupled as shown by the continuous shift of intensity from the upper to the lower branch observed[6,7] in both compounds.

As a result of this unusual coupling, broad soft phonons are observed in both compounds just above $T_c (\cong 15$ K) (see Fig. 4 of Ref. 7). Below T_c, a sharp peak near 4 meV, followed by a broad shoulder in the high-energy side, is observed in both compounds (see Fig. 4 of Ref. 7). Simulations demonstrated that the broad shoulder is not due to the folding of the resolution ellipsoid with the steep dispersion of the phonon branches. Furthermore, the measured[4,7] room-temperature phonon frequencies, up to 40 meV, were assigned to the various branches by comparing the measured intensities with calculations based on Born–von Kármán force-constant models. These calculations demonstrated that the room-temperature dispersion curves can be fitted quite well with force-constant models and that no other branches of the same symmetry are close to the branches under study.

To better understand the nature of the broad soft phonons just above T_c and the origin of the sharp phonon peak observed in both compounds below T_c, we performed a systematic study, between 4 and 620 K, of these anomalous phonons in both compounds. In this paper we report the results of these experiments.

II. EXPERIMENTAL DETAILS

The measurements were performed on single crystal samples of $LuNi_2B_2C$ and YNi_2B_2C grown at the Ames Laboratory. The crystals were platelets (1–2 mm thick with a plate area of 0.5–1 cm^2) with the c axis perpendicular to the

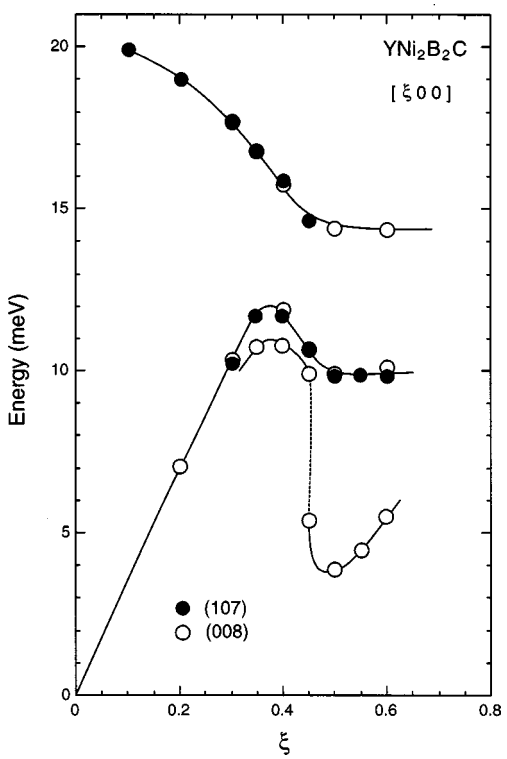

FIG. 1. $\Delta_4[\xi 00]$ branches of YNi_2B_2C at 150 K. Solid [open] circles denote data obtained around the (107) [(008)] reciprocal lattice point.

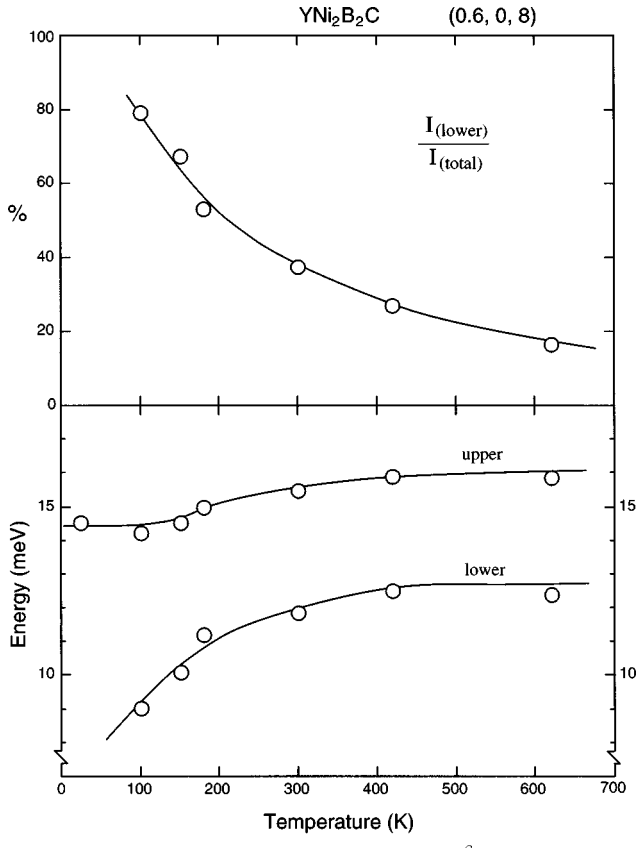

FIG. 2. Lower panel: temperature dependence of the frequencies of the lower and upper (0.6 0 0) modes of YNi_2B_2C. Upper panel: temperature dependence of the ratio of the intensity of the lower mode to the total intensity of the two modes.

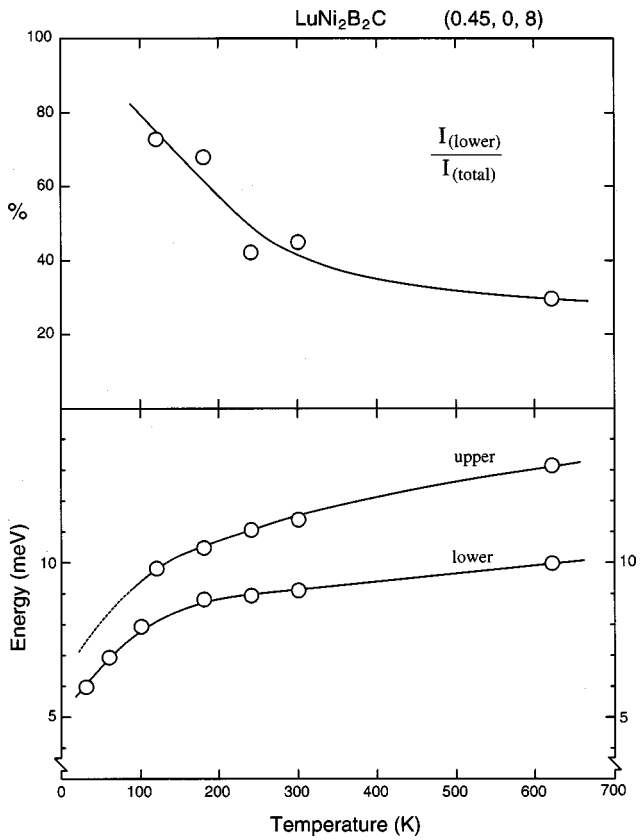

FIG. 3. Lower panel: temperature dependence of the frequencies of the lower and upper (0.45 0 0) modes of $LuNi_2B_2C$. Upper panel: temperature dependence of the ratio of the intensity of the lower mode to the total intensity of the two modes.

plate surface. The 300 K lattice parameters are $a=3.464$ Å, $c=10.623$ Å for $LuNi_2B_2C$ and $a=3.526$ Å, $c=10.547$ Å for the YNi_2B_2C compound.

The neutron-scattering experiments were performed using the variable incident neutron energy triple-axis spectrometers H-8, at Brookhaven National Laboratory, HB3 at Oak Ridge National Laboratory (ORNL), and the fixed initial energy triple-axis spectrometer HB1A at ORNL Pyrolitic graphite (PG), reflecting from the (002) planes, was used as monochromator and analyzer and a PG filter was used to minimize higher-order contamination. The measurements on the variable incident neutron energy spectrometers were performed either with neutron energy loss and fixed final neutron energy (E_f) of 14.7 and 30.5 meV or with energy gain and a fixed incident neutron energy (E_i) of 30.5 meV. The high-temperature measurements were performed on the HB1A spechrometer with neutron energy gain and fixed E_i at 14.7 meV. Most of the data were collected with a collimation of 40-40-40-40 minutes of arc in the standard positions on the spectrometers.

III. EXPERIMENTAL RESULTS

The detailed temperature dependence of the frequencies of the two Δ_4 branches, in the vicinity of $\xi=0.5$, was determined by measurements at (0.6 0 8) in the Y and at (0.45 0 8) in the Lu compound. The measured frequencies of the two modes and the ratio of the intensity of the lower to the total intensity of the two modes are plotted in Figs. 2 and 3. It can be seen (Figs. 2 and 3) that in both compounds the frequency of the lower mode decreases with decreasing temperature and intensity is transferred from the upper to the lower mode. It is important to notice that in the Y compound (Fig. 2) the frequency of the lower mode decreases much faster than that of the upper mode, especially below 300 K, and as a result the two branches separate in the vicinity of $\xi=0.5$ as the temperature is lowered. In the Lu compound (Fig. 3), on the other hand, the two branches approach each other in the vicinity of $\xi=0.5$ with decreasing temperature.

In the vicinity of and below T_c, measurements performed with optimized instrumental conditions and much longer counting times per point than in our previous study (Fig. 4 of Ref. 7), established that the broad peak observed just above T_c in both compounds corresponds to the acoustic mode. This broad acoustic phonon profile is abruptly changed into a sharp peak with a broad shoulder at the higher-energy side as the temperature is lowered below T_c. The width and position of the sharp peak remain practically unchanged below T_c, and its intensity decreases with increasing temperature in the vicinity of T_c (Fig. 5 of Ref. 7). It should be pointed out that in the present measurements the optical mode in the Y compound was visible down to 4 K (Fig. 2). In the Lu compound the intensity of this mode was too weak and its frequency too close to that of the acoustic mode to be observed below a temperature of approximately 150 K (Fig. 3).

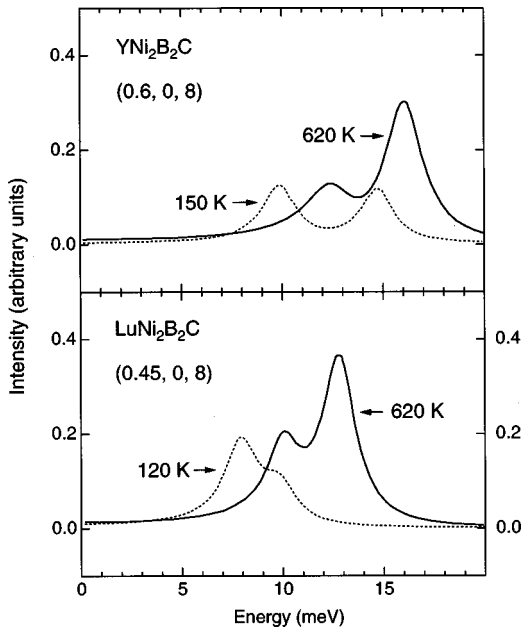

FIG. 4. Phonon profiles of YNi_2B_2C [at (0.6 0 8)] and $LuNi_2B_2C$ [at (0.45 0 8)] evaluated using the coupled-mode model (see text).

IV. DISCUSSION

The experimental results show that above T_c the temperature dependences of the frequencies of the lower Δ_4 mode in the vicinity of the Fermi surface nesting vector and the intensity transfer from the upper to the lower mode upon cooling are very similar in the two compounds. The key difference in the behavior of these modes above T_c is that in the vicinity of the nesting vector the two branches approach each other upon cooling in the Lu compound, whereas they separate in the Y compound.

We have shown previously[7] that the phonon profiles above T_c in $LuNi_2B_2C$ can be described in terms of a simple coupled-mode model which was used[10] successfully to explain the asymmetric phonon profiles observed in ferroelectric $BaTiO_3$. The observation that upon cooling the $LuNi_2B_2C$ Δ_4 branches approach each other, whereas in the Y compound separate, with approximately the same intensity transfer from the upper to the lower branch (compare the $I_{\text{lower}}/I_{\text{total}}$ curves in Figs. 2 and 3), suggests that by simply changing the sign of the coupling constant the coupled-mode model could describe as well the phonon profiles above T_c in YNi_2B_2C. This is illustrated in Fig. 4 where calculated phonon profiles for Lu (at 620 and 120 K) and Y (at 620 and 150 K) are plotted. It can be seen that these profiles are very similar to those observed experimentally. Thus the phonon profiles above T_c in both compounds can be described in terms of a coupled-mode model.

Several theoretical approaches[11–13] have been proposed to explain the dramatic change of the acoustic phonon profiles of the compounds upon entering the superconducting state. Allen *et al.*[12] follow the usual BCS approach for a three-dimensional quasi-isotropic system. Kee and Varma[11] and Maksimov and Karakozov,[13] on the other hand, assume the existence of a Fermi surface nesting vector $\vec{\xi}_m \cong (0.5\ 0\ 0)$, as obtained by band theoretical calculations.[13] The calculated phonon profiles[11,12] are in qualitative agreement with the experimental results. The theoretical calculations, on the other hand, predict that the energy of the sharp peak must follow the temperature dependence of the superconducting gap 2Δ, whereas the experimental observations show that the energy of this peak is practically temperature independent below T_c (see Fig. 5 of Ref. 7). The intensity of this sharp peak does decrease with increasing temperature, but it goes to zero at T_c more abruptly than the superconducting gap.

It is clear that a detailed and fundamental understanding of the phonon spectra in these compounds, both above and below T_c, must wait future first-principles theoretical calculation. Of particular importance here is to elucidate the connection between the intensity variation of the sharp peak below T_c and the superconducting properties of these compounds.

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[1] C. Stassis and A. Goldman, J. Alloys Compd. **250**, 603 (1997) and references therein.
[2] J. Lynn, J. Alloys Compd. **250**, 552 (1997) and references therein.
[3] J. Y. Rhee, X. Wang, and B. N. Harman, Phys. Rev. B **51**, 15 585 (1995).
[4] P. Dervenagas, M. Bullock, J. Zarestky, P. Canfield, B. K. Cho, B. N. Harmon, A. Goldman, and C. Stassis, Phys. Rev. B **52**, R9839 (1995).
[5] K. Kawano, H. Yoshizawa, H. Takeya, and K. Kadownki, Phys. Rev. Lett. **77**, 4628 (1996).
[6] C. Stassis, M. Bullock, J. Zarestky, P. Canfield, A. Goldman, G. Shirane, and S. M. Shapiro, Phys. Rev. B **55**, 8678 (1997).
[7] M. Bulock, J. Zarestky, C. Stassis, A. Goldman, P. Canfield, Z. Honda, G. Shirane, and S. M. Shapiro, Phys. Rev. B **57**, 7916 (1998).
[8] F. Gompf, W. Reichardt, H. Schober, B. Renker, and M. Bschgeister, Phys. Rev. B **55**, 9058 (1997).
[9] I. K. Yanson, V. V. Fisun, A. G. M. Jansen, P. Wyder, P. C. Canfield, B. K. Cho, C. V. Tomy, and D. McK. Paul, Phys. Rev. Lett. **78**, 935 (1997).
[10] J. Harada, J. D. Axe, and G. Shirane, Phys. Rev. B **4**, 155 (1971).
[11] H. Y. Kee and C. M. Varma, Phys. Rev. Lett. **79**, 4250 (1997).
[12] P. B. Allen, V. N. Kostur, N. Takesue, and G. Shirane, Phys. Rev. B **56**, 5552 (1997).
[13] E. G. Maksimov and A. E. Karakozov, Solid State Commun. **107**, 353 (1998).